

1 The Origin of Blue Coloration in a Fulgurite from Marquette, Michigan

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22 **Abstract**

23 Few rocks or minerals display a blue hue; this is especially true for natural
24 glasses. **Due to the specific internal fragile structures**, glasses are already uncommon
25 compared to crystalline rocks, the rarity of blue glass makes a blue fulgurite found in
26 Marquette Michigan worthy of further examination. We study the morphology and the
27 chemical and structural characteristics of the blue fulgurite using Raman
28 spectroscopy, X-Ray Fluorescence analysis, Election Microprobe Analysis, and
29 Transmission Election Microscopy. We use the experimental results to compare the
30 fulgurite with another blue natural glass, zhanmanshinite, an impact glass named after
31 an exposed impact crater in Kazakhstan, and to evaluate the support for several
32 hypotheses of the origin of the blue color in the fulgurite.

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34 Key Words: fulgurite, blue glass, immiscible melt, cristobalite

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361. Introduction

37 Coloration within rocks and minerals may be due to inherent elemental causes
38 (idiochromatic), from trace elements (typically rare earth or transition metals) present
39 as impurities (allochromatic), or by non-chemical causes (pseudochromatic).
40 Similarly, synthetic glasses may be colored using major components, trace additives,
41 or physical effects (e.g., Stookey 1949, Stroud 1971, Puche-Roig et al. 2008). Studies
42 of the coloration of natural glasses is rather limited in contrast to minerals (see
43 Zolensky and Koeberl 1991, Ma et al. 2007) due to the general paucity of glasses
44 produced by natural phenomena and their short-term persistence as glass is
45 susceptible to devitrification reactions, typically caused by water.

46 While terrestrial glasses are rare, there are several types of natural events that
47 form glasses on Earth (O'Keefe, 1984). Natural glasses can form via high
48 temperatures that are caused by friction during seismic events, rapid cooling of lava,
49 or during high energy events such as a meteor impact/cloud-to-ground lightning
50 strike. Some examples of natural glasses are obsidian, pumice, scoria, and tachylite
51 from volcanic sources. Generally, pseudotachylites are formed by brittle or brittle-
52 ductile deformation of rock during a seismic event (Philpotts, 1964), sometimes an
53 impact. Tektites are formed by the aerodynamic cooling of molten material ejected by
54 impact (Carron & Jun, 1961).

55 Fulgurites occur on crustal areas of the Earth and are formed by electric
56 discharge, typically when lightning strikes sand, soil, or rock during cloud-to-ground

lightning (Yavuz & Can, 1999, Spoonholz, 2004). With forty-five lightning strikes occurring each second on Earth (Christian et al., 2003) and more than 90% of those strikes occurring over crustal land mass (Altaratz et al., 2003; Williams et al., 2004; Lay et al., 2007), and with about 25% of the lightning occurs as cloud-to-ground lightning, as many as 10 fulgurites may be formed on Earth every second (Pasek & Block, 2009). As fulgurites can form over any area, fulgurites should be as chemically diverse as the sand, soil, and rock in which they form. A fulgurite's morphology (structure) traces the primary path of the electric discharge as the rock, soil, or sand is melted and vaporized through the central plasma channel. The chemical composition of the fulgurite is dependent on the parent material as the energy heats and often melts the minerals present.

Natural fulgurites are formed when cloud-to-ground lightning is the source of the discharge, whereas artificial fulgurites are formed when a high voltage or high current from the breakdown of electric circuits is the source of the discharge (Lowry, 1975; Kumaziki et al., 1997; Bidin et al., 2018; Elmi et al., 2018; Pasek and Pasek, 2018,). The blue fulgurite used in this study from Marquette, Michigan, USA (Figure 1) is a fulgurite formed in 2014 when lightning struck a tree, causing it to fall over against two high voltage power lines, with the current then travelling through the tree into sandy-clayey soil along what was apparently a tree root for three hours (Brzys 2014). This fulgurite bears characteristics of both natural and artificial sources as per the classification scheme of Pasek and Pasek (2018). The current forming the

Marquette fulgurite was prolonged, which resulted in a long length, large thickness, and blocky, glassy matrix.

[FIGURE 1]

The Marquette fulgurite is similar in appearance to blue zhamanshinite, which is an impact melt glass named after an exposed impact crater in Kazakhstan. The blue glass is found with a variety of other impact glasses consisting of other colors, as well as tektite-like glasses that are termed irghizites (Koeberl, 1986). The 900,000 (\pm 100,000)-year old crater has a diameter of 13 kilometers with a bottom width of 6.5 kilometers (Masaitis, 1999). The crater is geologically complex with a center of loess and lake sediment. The rim is composed of metamorphic crystalline rocks which include both volcanic and sedimentary series cut by ultrabasic veins (Koeberl, 1986). Furthermore, this crater may be the source of the Australasian tektites, which have an extensive strewn field (Koeberl 1988a, Glass 1990). Generally, the natural glass found in the crater area is referred to as “zhamanshinite.” While the existence of the blue glass was first noted by Florenskij and Dabizha in 1981, it was not until Koeberl (1986) chemically examined the glass that the glass was distinguished from other impact glasses. By doing so, Koeberl (1988b) provided major and trace chemical data for the blue zhamanshinites. Zolensky and Koeberl (1991) further examined the morphology and major chemical composition of the blue zhamanshinites, described immiscible glass inclusions, and discussed the role of phosphate inclusions as the

cause of the Rayleigh scattering that results in the blue coloring of the glass. This scattering is akin to scattering that results in atmospheric coloration (Bucholtz, 1995).

Previous researchers have presented classification schemes for specific tektites and impact glasses found at the Zhamansin crater, and for fulgurites. Koeberl (1986) classified glasses into irghizites and into Si-rich (70-80 wt. % SiO₂) or Si-poor (less than 55 wt. % SiO₂) impact glasses (Koeberl, 1988b). Pasek et al. (2012) classified fulgurites by morphology and divided the fulgurites into four categories. Type I consists of the sand fulgurites with thin glass walls; Type II contains the clay fulgurites with thick, melt-rich walls; Type III encompasses the caliche fulgurites with thick, glass-poor walls; and lastly, and Type IV includes the rock fulgurites with glass walls surrounded by unmelted rock. This fulgurite classification was expanded by Pasek and Pasek (2018) to fulgurites forming from unnatural discharge sources, and from those forming in manmade materials. We apply this prior work to this (thus far) unique blue fulgurite.

Because blue zhamanshinite is the only other example of blue color volcanic glass that ever found until now. We employ Raman spectroscopy, X-Ray Fluorescence (XRF) analysis, Election Microprobe Analysis (EMPA), and Transmission Election Microscopy (TEM) to determine the major and minor chemical composition of the Marquette fulgurite and to examine the blue fulgurite's microscopic morphology. Furthermore, we utilized these experiment data to compare

with blue zhamanshinite (Zolensky and Koberl 1991) to distinguish if these two similar blue glass have same characters with blue hue.

We also use the morphology and chemical composition results to classify the fulgurite according to prior fulgurite classification schemes, and same, to compare the fulgurite with the data from Zolensky and Koeberl's (1991) earlier analysis of the blue zhamanshinite. We hypothesize that the origin of the blue coloration in this fulgurite is pseudochromatic, due to spherical inclusions in the host glass that create conditions conducive to the scattering of light, yielding a strong blue color. The similarities and differences between the blue fulgurite and blue zhamanshinites may assist in determining some of the P-T-t (Pressure- Temperature- time) features of both discharge and impact cratering events.

2. Methods and Materials

Samples of blue fulgurite glass, and a sample of the parent soil (from the finder via eBay, located at Marquette, Michigan) were analyzed (Figure 2). The fulgurite was found near the south side of Lake Superior(46°32'47"N,87°24'24"W). A thick section was prepared for analysis. The thin section was mounted on a glass slide using epoxy and polished as per standard methods.

[FIGURE 2]

137 Raman point microanalysis of the fulgurite was used to identify minerals in
138 thick section, using an Enwave Opt. Inc. (Model No. EZI-785-A2). The Raman
139 microscope is a Leica DM300 microscope equipped with three objective lenses
140 ($\times 4/0.1$ NA, $\times 10/0.25$ NA and $\times 40/0.65$ NA). The spectra were processed using
141 Crystal Sleuth freeware (Laetsch & Downs, 2006) and compared to relevant spectra
142 from the RRUFF database (Lafuente et al, 2015).

143 Samples of the glass and the parent soil were analyzed by X-Ray Fluorescence
144 (XRF) at Hamilton College (NY) on a Bruker AXS S8 Tiger Wavelength Dispersive
145 X-Ray Fluorescence (WDXRF). These samples were also analyzed using Electron
146 Probe Micro-Analysis (EMPA) to determine the major elements of the fulgurite.
147 Electron Probe Micro-Analysis was performed on a JEOL 8900R Super Probe
148 (Florida International University) after samples were carbon coated, and samples were
149 standardized with comparison to EMS-1 Obsidian (Price & Pichler, 2006). In this
150 experiment, we remotely probed the fulgurite across a cross section to measure the
151 weight percentages of the major elements in the sample, averaging these points to
152 determine “typical” composition.

153 Structures on the submicron scale were investigated using a Tecnai G2 F20
154 TWIN 200 kV / FEG Transmission Electron Microscope (TEM) with a Gatan
155 UltraScan 4000 (4k x4k) CCD camera at University of Florida. Bright Field
156 Transmission Electron Micrograph analysis (BFTEM) and a High Angle Annular
157 Dark Field Scanning Transmission Electron Micrograph analysis (HAADF-STEM)

specifically were chosen to determine the shape of several nano-scale structures. In addition, Energy Dispersive X-Ray Spectrometry (EDS) was utilized to map major elements inside the inner glass (EDAX GENESIS XM2).

3. Results

The blue fulgurite was formed as a massive cylindrical shape with multiple tubular chambers. A majority of the blue glass-bearing tubes are massive (75% glass width), with little internal void (0-25%)(figure?), consistent with an artificial electric discharge source (Pasek and Pasek 2018). In contrast, the less intensely-blue hued samples have larger void proportions (27%), implying these may have been formed during the initial lightning strike. The fulgurite has a hardness of about 4.5 on the Mohs Hardness Scale. The sample does not display cleavage or magnetic attraction. Under fluorescent light, the sample changes color from blue to purple. Furthermore, a thin section changes color to green as it is illuminated from behind with plane polarized light. The soil that was collected by the finders “adjacent” to the fulgurite was a sample of sandy-clay sediment.

The average composition of the fulgurite glass is provided as table 1 & 2, based on microprobe and XRF data. Comparison of this data to other fulgurites and zhamanshinites in the literature is provided as a ternary diagram (Figure 3). A compositional comparison to the soil suggests the fulgurite and adjacent soil are not

compositionally related, or suggests significant fractionation occurred during the heating event.

[Table 1]

[Table 2]

[Figure 3]

Per Figure 3, while this fulgurite is similar to the blue zhaminshinites in color and similar to type II fulgurites in morphology, this fulgurite is chemically distinct from either group (Figure 3), as it is more Fe-, Mg-, and Ca-rich.

Raman point analysis of the fulgurite samples generally showed the major material to be highly amorphous, and with little to no Raman scattering peaks. However, a large white mineral grain was found in the sample which was determined through Raman analysis to be cristobalite (SiO₂). Furthermore, blackened material was also found on the sample which is most likely charred wood. Raman characterization of this charcoal material indicated that much of the material had been heated to the point that it was highly aromatic, but also very disordered, given the difference between the D and the G bands observed by Raman (Figure 4).

[FIGURE 4]

TEM analysis of the fulgurite reveal the presence of abundant nanospheres (~100 nm in diameter, Fig 5&6). EDS mapping of spheres at this scale showed that the

major element inside the spheres was Si (Fig 7), and were in general much lighter in elemental composition than the glass groundmass.

[Figure 5]

[Figure 6]

[Figure 7]

4. Discussion

As per the Pasek et al. (2012) classification scheme, this fulgurite is a Type II fulgurite based on its morphology. We consider now the origin of the blue coloration of this fulgurite. Blue coloration within fulgurites has not been reported previously to date (this is the only known example). Most Type I (sand) fulgurites are white to beige, reflecting the coloration of the original sand. Type II fulgurites are more diverse in color, with typical samples ranging from brown to gray-black to bottle-green. Given the uniqueness of this coloration, we consider the possibility that the fulgurite may have formed through an unusual heating event or environment. We then consider the origin of the blue coloration as due to chemical interactions.

4.1 Origin of the Fulgurite

The fulgurite as described by the finders would be characterized as a natural and artificial fulgurite (describing the source of the discharge). The presence of cristobalite supports a longer formation timescale than typical fulgurites formed solely by lightning, as the conversion of quartz to cristobalite requires elevated temperatures that persist for minutes to hours (Pasek and Pasek 2018). Although Elmi et al. (2017) (Crespo et al., 2009?) report cristobalite within a fulgurite from Italy, the formation temperature of these fulgurites appears to have been moderated by the combustion of lichen, which may have allowed for elevated temperatures to have persisted for long enough to convert quartz to cristobalite (or the fulgurite may have a partially artificial origin). Beyond this one occurrence of cristobalite within a natural fulgurite, cristobalite is surprisingly absent from the mineralogy of natural fulgurites. The longer persistence of high temperatures during formation of this fulgurite may have allowed for the separation of the matrix into two immiscible components. This may indicate why no wholly natural fulgurites have been reported with a strong blue hue.

When we compare the chemical compositions of the blue fulgurite to that of the Type II fulgurites (Pasek et al., 2012) we see that this fulgurite is not chemically similar. The presence of cristobalite and graphite in the fulgurite instead of lechatelierite—amorphous SiO_2 —indicates this fulgurite was not formed in the same manner as a typical Type II fulgurite. The formation of cristobalite limits the temperature reached to less than roughly 2000 K, the melting point of SiO_2 (Breneman & Halloran, 2014). This indicates slower heating as opposed to the rapid

heating at high temperatures typical of Type II fulgurites (Pasek and Pasek, 2018).
The presence of multiple chambers where tree roots would be expected also supports
the conclusion that the lightning struck the tree and not the ground, as does the lack of
melted conducting wire (also an indicator of origin per Pasek and Pasek 2018).

The fulgurite is otherwise not compositionally distinct from other Type II
fulgurites, though it is slightly poorer in SiO₂, and richer in CaO and MgO than most
Type II fulgurites (Fig. 3)(Crespo et al.,2009; Pasek et al., 2012). Morphologically
the fulgurite is consistent with a Type II fulgurite, though some of the more massive
pieces match well with artificial fulgurites as per the scheme in Pasek and Pasek
(2018). Pasek and Pasek (2018) do note that determining the origin of a type II
fulgurite based on morphology is less robust than finding melted conductor wire
and/or cristobalite.

4.2 Cause of the Blue Coloration

We evaluate the cause of the blue glass, considering whether the blue coloration is
due to the presence of specific compounds that modify the color of the glass (e.g.,
copper), or to other physical features. The bulk composition of the fulgurite is
similar with other fulgurites, which eliminates the possibility of idiochromaticity
being the cause for the unique color. In addition, Table 2 shows the amount of typical
abundances of key trace elements in blue fulgurite, blue Zhamanshinites, Australite

Tektites and Fulgurite of Torre de Moncorvo. The abundances of some special trace element that contribute blue color in minerals, like Co, Ni, Cu and Zn, are almost same amount in ppm. However, only blue fulgurites and blue zhamanshinites have blue color, and other two samples do not have special blue hue. This evidence can be considered that dislike azurite and malachite, the abundance of trace elements do not suggest any allochromatic effects in blue fulgurites .

We believe the blue coloration in blue fulgurites is due to pseudochromatic effects, namely Rayleigh scattering by nanoscale spherules within the glassy matrix, as demonstrated by the TEM images (FIG 10). Thus, the blue coloration within the Marquette fulgurite is likely akin to the blue coloration of blue Zhamanshinites as described by Zolensky and Koeberl (1991): nanospheres formed from separation of immiscible components within a melt. The blue zhamanshinites also had spherules with a similar size (~100 nm). Unlike the blue zhamanshinite samples, the spherical inclusions within the Marquette fulgurite are not rich in phosphorus but instead are silica-rich with a lighter average atomic number. While these impurities could impart some color to the fulgurite, none account for the blue color. It is more probable these inclusions create the physical conditions conducive to Rayleigh scattering of light and give the fulgurite its blue color.

5. Conclusions

277 The intense blue coloration of the Marquette, Michigan fulgurite has only one
278 natural analog: the blue zhamanshinites. The coloration is likely due to the presence
279 of nanoscale spherules that scatter light. However, composition of the spherules
280 does not appear to be important in the scattering of light, as the spherules within the
281 zhamanshinite are more P-rich than the Si-rich fulgurite spherules. Instead, the size
282 and ubiquity of the spheres in the matrix is likely critical to providing a blue hue to
283 glass (Kawamura et al. 2017).

284 Although this investigation may seem to be narrow in focus, the investigation of
285 the Marquette, Michigan fulgurite may provide some important clues on the
286 Zhamanshin cratering event (and thereby cratering as a whole). Specifically, the
287 requirement for a longer duration heating event for the formation of a blue hue (3
288 hours) may suggest that the blue glass at Zhamanshin crater formed with the
289 persistence of high-temperature heat for hours. Shorter timescales appear to be
290 unable to generate the conditions necessary for the development of spherules and,
291 thereby, the immiscible melt and blue coloration, given that natural fulgurites thus far
292 have not demonstrated consistent blue coloration. This work falls within a broader
293 theme of investigating parallels between lightning-formed materials and impact
294 materials, such as elemental reduction (Essene and Fisher 1986, Pasek 2008, Pasek
295 and Block 2009) and the potential for forming shocked quartz by lightning (Carter et
296 al. 2010, Ende et al. 2012, Giere et al. 2015, Chen et al. 2017).

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Table 1. Major element abundances (in wt%) in the blue fulgurite and associated soil sample. Compared with Zolensky& Koeberl (1991). Note: “BDL” stands for Below Detection Limits and represents areas that were below the detection limits of instruments used in this study.

	EPMA	XRF	XRF	Zolensky, Koeberl, 1991
	Blue fulgurite	Blue fulgurite	Soil	Blue Zhamanshinite
SiO ₂	62.98	62.70	75.14	74.0
Al ₂ O ₃	7.65	7.32	6.43	11.02
FeO*	2.7	3.41	1.42	4.47
CaO	16.06	15.89	3.29	4.08
MgO	9.90	7.36	1.37	1.33
K ₂ O	1.36	1.41	2.00	2.7
Na ₂ O	1.21	1.32	1.00	1.52
TiO ₂	0.15	0.35	0.19	0.58
MnO	0.09	0.09	0.04	0.10
Cr ₂ O ₃	0.02	BDL	BDL	BDL
ZrO ₂	0.03	BDL	BDL	BDL
P ₂ O ₅	0.15	0.13	0.08	0.80
Total	102.3	99.98	90.97	100.6

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515 **Table 2.** Trace element abundance (in ppm) of blue fulgurite and soil sample.
 516 Compared with Koeberl (1988), **Taylor and McLennan(1979)**, **Martin Crespo et al.**
 517 **(2009)**. Note: “BDL” is below the detected limits of the instruments used

Minor element	Blue fulgurite glass	Blue fulgurite soil	Zh 31/6A	Zh31/6 B	BZ 8601	Australite tektites	Fulgurite of Torre de Moncorvo
Sc	BDL	BDL	11.2	9.0	10.3	13	3.33
V	54	22	BDL	BDL	BDL	83	8.83
Cr	BDL	BDL	167	111	170	145	17.17
Co	BDL	BDL	24	14	14.5	25	BDL
Ni	18	7	<300	<200	<200	105	3.63
Cu	25	14	BDL	BDL	BDL	6.5	19.10
Zn	26	44	BDL	BDL	BDL	BDL	57.53
Ga	7	9	20	18	15	BDL	18.23
As	BDL	BDL	2.5	3.2	5.3	BDL	6.38
Se	BDL	BDL	BDL	BDL	BDL	BDL	0.4
Br	BDL	BDL	<1	<1	<1	BDL	0.3
Rb	36	46	74	89	98	BDL	302.17
Sr	156	176	180	220	150	BDL	80.97
Y	13	6	BDL	BDL	BDL	31	12.97
Zr	138	133	450	600	210	264	60.72
Nb	4.7	2.2	BDL	BDL	BDL	18.7	10.75
Mo	BDL	BDL	BDL	BDL	BDL	0.3	0.13
Sn	BDL	BDL	BDL	BDL	BDL	1.3	33.83
Sb	BDL	BDL	1	1	1.18	BDL	3.87
I	BDL	BDL	BDL	BDL	BDL	BDL	0.90
Cs	2	0	4.8	4.8	7.97	5.7	28.47
Ba	317	505	305	240	330	356	249.87
La	14	20	27.9	21.2	26.2	36.9	6.92
Ce	30	25	70.1	54.7	64.8	78.6	23.7
Nd	15	11	32	26.5	29	35	8.93
Sm	BDL	BDL	6.58	4.60	5.70	6.1	7.10
Eu	BDL	BDL	1.36	1.09	1.41	1.17	BDL
Gd	BDL	BDL	5.6	5.0	5.4	5.34	BDL
Tb	BDL	BDL	1.0	0.79	0.95	0.84	BDL
Dy	BDL	BDL	4.80	3.70	5.7	5.17	BDL
Yb	BDL	BDL	3.36	2.58	3.95	2.8	BDL
Lu	BDL	BDL	0.53	0.37	0.49	BDL	BDL
Hf	7.3	5.0	6.4	6.3	6.17	7.1	7.10
Ta	BDL	BDL	0.8	0.7	BDL	BDL	1.87
W	BDL	BDL	1	<2	<3	0.39	6.88
Au	BDL	BDL	0.01	<0.04	0.022	BDL	BDL
Tl	BDL	BDL	BDL	BDL	BDL	BDL	1.85

Pb	5	22	BDL	BDL	BDL	5	49.92
Bi	BDL	BDL	BDL	BDL	BDL	BDL	5.34
Th	5	9	7.23	7.29	8.07	13.7	4.15
U	2	1	3.1	3.1	3.18	2.1	14.57

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521 **Figure 1.** Fulgurite in Marquette, Michigan (Brzys 2014), with general overview (a)
522 and in situ (b&c) pictures.

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524 **Figure 7.** TEM images of (a) spherules within the fulgurite, and (b) corresponding EDS
525 elemental map, with blue corresponding to Si, green to Mg, and red to Ca.

526

527 **Figure 2.** Bulk sample of blue fulgurite, showing cross-sectional (a & c) and lateral
528 views (b)

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530 **Figure 3.** Ternary Diagram, which displays the blue fulgurite and other Type I, II, III,
531 and IV fulgurites from Pasek et al., 2012. Sheffer 2007, Crespo et al., 2009, Elmi et al.,
532 2017.

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534 **Figure 4.** Laser Raman spectroscopy figures of the host minerals of blue glass, with
535 a) cristobalite (RRuff denotes Raman spectrum of a cristobalite standard) and b)
536 charred root that has been partially graphitized (RRuff denotes Raman spectrum of a
537 graphite standard).

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539 **Figure 5.** Bright Field Transmission Electron Microscopy (BFTEM) image of the
540 fulgurite sample. In BFTEM, lighter contrasts correspond to lighter Z elements.

541 **Figure 6.** High Angle Annular Dark Field Scanning Transmission Electron
542 Microscopy (HAADF-STEM) image of the fulgurite. With DFTEM, darker
543 contrasts correspond to lighter Z elements.

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545 **Figure 7.** TEM images of (a) spherules within the fulgurite, and (b) corresponding
546 EDS elemental map, with blue corresponding to Si, green to Mg, and red to Ca.